Journal of Hazardous Materials 369 (2019) 691–698

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Distribution of plastic polymer types in the marine environment; A meta-analysis

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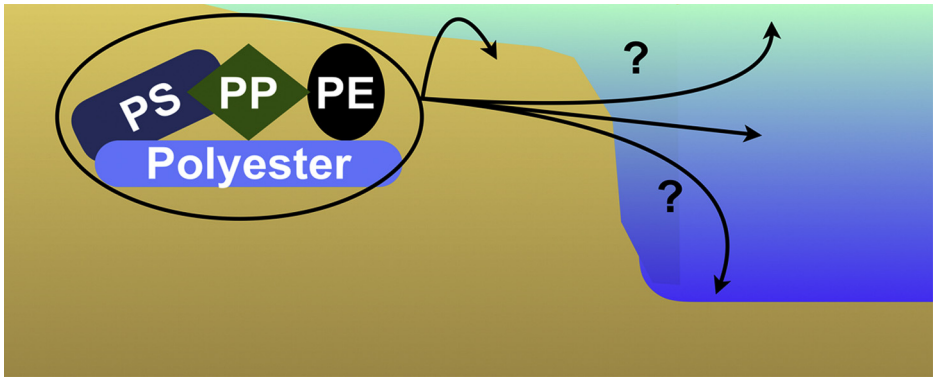
GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Microplastics
Marine ecosystems
Marine plastic debris
Polymer type
Environmental sinks

ABSTRACT

Despite growing plastic discharge into the environment, researchers have struggled to detect expected increases of marine plastic debris in sea surfaces, sparking discussions about "missing plastics" and final sinks, which are hypothesized to be coastal and deep-sea sediments. While it holds true that the highest concentrations of plastic particles are found in these locations (10^3-10^4 particles m^{-3} in sediments *vs.* 0.1–1 particles m^{-3} in the water column), our meta-analysis also highlights that in open oceans, microplastic polymer types segregated in the water column according to their density. Lower density polymers, such as polypropylene and polyethylene, dominated sea surface samples (25% and 42%, respectively) but decreased in abundance through the water column (3% and 2% in the deep-sea, respectively), whereas only denser polymers (*i.e.* polyesters and acrylics) were enriched with depth (5% in surface seawater *vs.* 77% in deep-sea locations). Our meta-analysis demonstrates that some of the most abundant and recalcitrant manufactured plastics are more persistent in the sea surface than previously anticipated and that further research is required to determine the ultimate fate of these polymers as current knowledge does not support the deep sea as the final sink for all polymer types.

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https://doi.org/10.1016/j.jhazmat.2019.02.067
Received 11 November 2018; Received in revised form 4 February 2019; Accepted 20 February 2019
Available online 21 February 2019

1. Introduction

Plastics benefit human society in numerous ways, such as improved consumer health and product durability or reduced CO_2 emissions with lightweight materials [1], but in recent years plastic has been identified as a widespread and recalcitrant pollutant in aquatic environments. It was estimated that over 8 million tons of plastic enter the oceans annually [2], and plastic is now found in all major oceanic gyres [3], polar seas [4] and deep sea sediments [5]. In the environment, plastic is known to deteriorate and fragment [6,7] therefore occurring in a wide range of sizes [8,9], for which standardized categories have very recently been proposed: macro- (≥1 cm), meso- (1–10 mm) and micro- (1–1000 µm) and nanoplastics (1–1000 nm; [10]), with smaller particles being numerically most prevalent on sea surfaces [3]. In the water however, degradation is slowed down by lower temperatures and limited UV penetration [7], leading plastic debris to persist and accumulate [11,12].

The global distribution of marine plastic debris and its broad range of particle sizes imply interactions with marine fauna at all trophic levels [13–18]. Thus far, much of the science has focused on the severity of impacts, while the probability of encounter received less attention. A complete understanding of risk however, can only be achieved by evaluating the availability of microplastics to biota [19] and potential release of additives embedded in them [20], which requires knowledge about the major sinks for plastic debris in marine ecosystems. Severity of ecological impacts will presumably be higher at plastic sink sites, which can be found through a better understanding of maritime plastic transportation [21,22].

Despite growing plastic production and discharge into the environment, researchers have struggled to detect predicted increases of small microplastic (< 1 mm) in sea surfaces, which has sparked discussion about possible sinks for marine plastic debris in deep sea sediments [3,23,24]. Various plastic polymer types have higher densities than seawater ($\rho > 1.02\ g\ cm^{-3}$, Table S1) which should logically lead to sinking, and in fact, plastics are plentifully found in the deep sea [5,25]. Even the most abundantly manufactured polymers, *i.e.* polyethylene (PE), polypropylene (PP) and some forms of polystyrene (PS) which are less dense than seawater, can sink when biofouled because of the increased density [26,27], or when included in fecal pellets after ingestion and marine snow [28–30]. Some factors in these processes remain uncertain though, such as the extent to which oligotrophic marine systems support biofilms large enough to cause sinking, or the effect of de-fouling and particle disaggregation, which would ultimately cause lower density plastic debris to resurface [27].

In light of uncertainties about the final sink for microplastic in aquatic environments, we conducted a meta-analysis in an attempt to identify patterns in the abundance of common synthetic polymer types in different aquatic zones. Investigating incidence of individual polymer types is now possible, owing to the routine implementation of spectroscopic methods to identify polymer types in environmental surveys. By focusing on polymer type, we demonstrate that relative abundance of specific synthetic polymers differs with sampled aquatic zone.

2. Methods

2.1. Eligibility criteria and search method

Literature on aquatic plastic debris was systematically reviewed considering only studies that clearly specified the aquatic zones that had been sampled, and in which the polymer type was identified.

Literature was searched using all databases in *Web of Science* and, as in Rochman et al. [31], the databases of the journals *Environmental Science & Technology* and *Marine Pollution Bulletin* due to their relevance in the field, yielding additional articles not found through *Web of Science*. The following Boolean search terms and modifiers were employed:

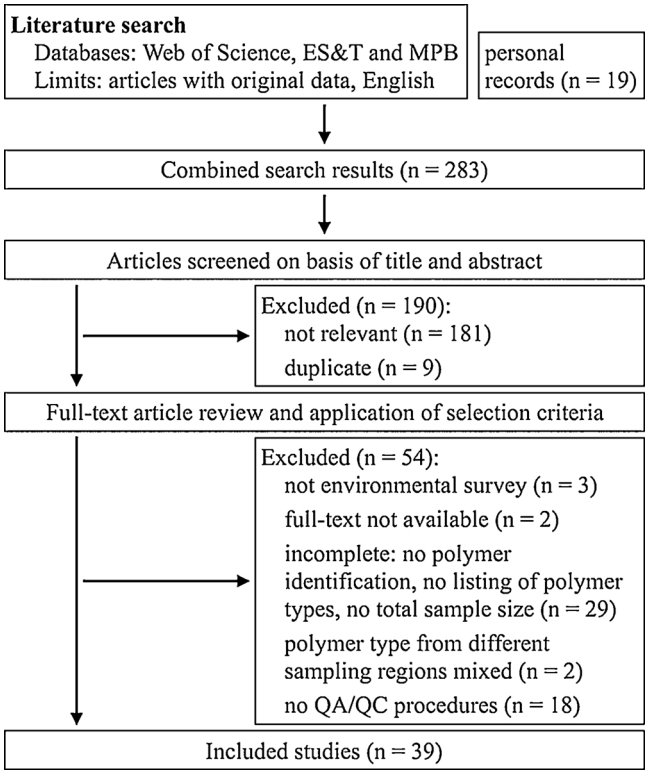

Fig. 1. Flow diagram of study selection. ES&T: Environmental Science & Technology; MPB: Marine Pollution Bulletin.

plastic AND debris AND environment* AND (FT$IR or Raman). The search included every available publication until March 2018, but was restricted to accepted, peer-reviewed publications in English with original data (Fig. 1).

2.2. Quality assessment and data extraction

Literature assessment was performed, in accordance with the predefined criteria described above. In a first step, publications were screened for relevance as environmental surveys by their title and abstract. In a second step, the materials and methods section of each publication was checked to assure that synthetic polymer types had been identified, and total sample size of the characterized polymer types was reported. If total sample size was missing, respective corresponding authors were contacted *via* email. Given the importance of controlling for contamination in microplastic research [32], studies were further checked for QA/QC procedures. Studies that failed to describe any type of control measures were no longer considered.

A sheet was developed for systematic data extraction. Data was recorded independently by two authors as follows: sample type (*i.e.* water or sediment), water type (*i.e.* marine or fresh), sampled zone (*i.e.* intertidal, subtidal, sea surface, water column, deep sea water (> 200 m depth) and deep sea sediment (> 200 m depth)), plastic extraction method (*i.e.* visual or density separation including employed density), maximum considered particle size, total sample size of characterized plastic particles, identified polymer types, and their respective relative abundance. Disagreements between reviewers during data extraction were resolved by consensus. In microplastic studies that did not report maximum particle size, a maximum particle size of 5 mm was assumed if such a definition was found in the introduction. In studies that reported microplastics from the environment as well as from biota, only microplastics from the free environment were considered. Separate entries were recorded for data from the same study if researchers sampled in different zones (*i.e.* depth related). Because the

present analysis focused on purely synthetic polymers, where relevant, total sample sizes and polymer proportions were adjusted to account for the exclusion of cellulose-based polymers, such as rayon, cellophane and cellulose acetate, which in some studies were included as artificial polymers. The original data and analysis script are available from the authors upon request.

Despite the focus on prevalence of polymer types in each sampled zone, we also considered reported plastic concentrations (particles m^{-3}). For studies that did not report particle concentrations by volume, these were calculated using total reported number of particles and total reported sampling volume. Polymer specific concentrations were then obtained based on respective polymer type prevalence. Absolute particle concentrations could be obtained or calculated from 20 studies.

2.3. Statistical analysis

The main outcome of interest in meta-regression models was the proportion of individual polymer types. The pooled prevalence of polymer types was calculated with arcsine square root transformed proportion data, because proportions of 0 commonly occurred. Potential bias at the level of polymer type was investigated *via* funnel plots and the significance of eventual biases was tested for using *Trim and Fill* followed by Egger's regression tests. Heterogeneity was assessed using the I^2 statistic *via* random effects models using the Paule-Mandel estimation method, which was previously found to be a better alternative for dichotomous data than other estimators [33]. Mixed effects meta-regression models were then employed for each of the studied polymer types separately, to test if the moderators sampling zone, debris size (*i.e.* studies reporting microplastic only or general plastic debris) and water type (*i.e.* fresh or saline) affected polymer type prevalence. To find the simplest possible model, explanatory variables were dropped stepwise and likelihood ratio tests were used to assess if the simplified models were significantly different from their previous versions.

To evaluate how methods to extract microplastics from sediments could have biased our results, such as an inflated prevalence of low-density polymers due to lower density extraction protocols, we estimated diversity of polymer types in sediment samples from intertidal and subtidal zones, as well as the effect of different extraction methods (*i.e.* solutions with density $\rho < 1.5\ g\ cm^{-3}$ and $\rho \geq 1.5\ g\ cm^{-3}$). For this, Shannon's diversity indices were calculated and, for ease of interpretation, converted to effective numbers of polymer types [34]. Standard errors were obtained *via* bootstrapping with 100 iterations. The threshold for density separation was set at $\rho = 1.5\ g\ cm^{-3}$, because all but one of the polymer types considered here, should be extracted *via* a solution of $\rho \geq 1.5\ g\ cm^{-3}$ (Table S1).

All analyses and plotting were performed in R (version 3.4.3, [35]), using the packages *meta* [36], *metaphor* [37], *vegetarian* [38], and *ggplot2* [39].

3. Results

3.1. Summary of included studies

The present meta-analysis draws data from 39 studies (Fig. 2). Initially, our literature search identified a total of 283 studies, but 190 were discarded after title and abstract screening, because they were not relevant to the topic (Fig. 1). After full-text review of the remaining 93 studies, another 53 were discarded mainly because authors did not report the polymer types and/or total sample size remained incomplete after contacting corresponding authors (n = 29), but also due to absent quality assurance and quality check (QA/QC) measures (n = 18). Further studies were excluded because they did not contain environmental data (n = 3), full text was not available (n = 2), or authors did not report polymer types from different sampling depths separately (n = 2). Missing total sample size or clarifications on the data were obtained from corresponding authors in seven of the final 39 studies considered [3,40–45].

Polymer type data for different sampling zones stemmed from 17 water surface studies (Fig. 3), 4 water column studies, 13 intertidal studies, 6 subtidal studies and three deep sea studies, including double entries for four studies. The subtidal studies were conducted in shallow regions with average sampling depths ranging between 6–59 m (median = 17.25 m), while the deep sea studies sampled at average depths of 2250 m, 2227 m, and 3496 m. Eight studies contained data from fresh water and estuarine sites, while 32 studies had marine data (including one study that sampled in both). In total, data on 24 different polymer types were recorded (Table S1), but due to sparsity of the data, meta-regression models were employed to study those that were most abundant: PE, PP, polystyrene (PS) and the group PP&A (polyesters, PEST; polyamide, PA; and acrylics). Despite that rare polymer types were not studied in greater detail, they were not subtracted from total particle counts; estimated relative abundance of PE, PP, PS and PP&A are therefore based on total characterized particle counts from all 24 polymer types.

3.2. Prevalence of polymer types in different aquatic zones

Pooled prevalence data confirmed that PE was the most abundant plastic type polluting aquatic environments with a predicted relative abundance of 23% (95% confidence interval 15–32%). The second most abundant polymer was of the group PP&A (20%; 95% confidence interval 11–32%), followed by PP (13%; 95% confidence interval 7–20%) and PS (4%; 95% confidence interval 2–9%). No significant publication bias was revealed by Egger's regression tests for any of the investigated polymer types (Fig. S1, PE: $Z = -1.59$, $p = 0.111$; PP: $Z = 1.08$, $p = 0.279$; PS: $Z = -0.48$, $p = 0.635$; PP&A: $Z = 1.42$, $p = 0.156$). Nevertheless, we found high heterogeneity ($I^2 = 98\%$) between studies at the level of all polymer types (Figs. 3 and S2-S5), indicating that the surveys did not share a common effect size.

Potential sources of heterogeneity were explored with meta-regressions using three categorical moderators, *i.e.* debris size, water type, and sampling zone. The simplest model for all polymer types, except PS, included a single significant moderator (*i.e.* sampling zone; see Table S2 for statistical details), indicating that of the moderators considered here, only "sampling zone" could explain part of the variability in polymer type prevalence observed between studies. PE and PP were relatively more abundant in surface samples (PE: 42% and PP: 25%) compared to water column- (PE: 9%, $Z = -2.55$, $p = 0.011$ and PP: 3%, $Z = -2.46$, $p = 0.014$), and intertidal samples (PE: 18%, $Z = 2.61$, $p = 0.009$ and PP: 5%, $Z = 2.66$, $p = 0.008$, Fig. 3). While the abundance of PE was also higher in surface water than subtidal sediments (11%, $Z = -2.29$, $p = 0.022$) and the deep sea (2%, $Z = -3.15$, $p = 0.002$), differences between PP prevalence in surface water compared to subtidal sediments and deep sea remained statistically insignificant (18%, $Z = -1.08$, $p = 0.279$; 3%, $Z = -1.87$, $p = 0.061$). In contrast, prevalence of PP&A was highest in deep sea- (77%) and water column samples (64%), and significantly lower in sea surface samples (5%, $Z = 3.519$, $p = 0.0004$; $Z = 3.344$, $p = 0.0008$ respectively). Despite accounting for sampling zone, heterogeneity remained very high throughout subgroups ($I^2 > 87\%$, Figs. S2-S5) indicating that further important moderators were missing from the models and that subgroups still did not share common effect sizes. Such additional variation can for instance stem from inconsistencies among sampling methodologies, which are reported in detail in the Supplementary Information.

3.3. Polymer concentrations in different aquatic zones

All common polymer types were most enriched in intertidal sediments ($\sim 10^3 - 10^4$ particles m^{-3}, Fig. 4). In surface waters, concentrations were four orders of magnitude lower than in intertidal

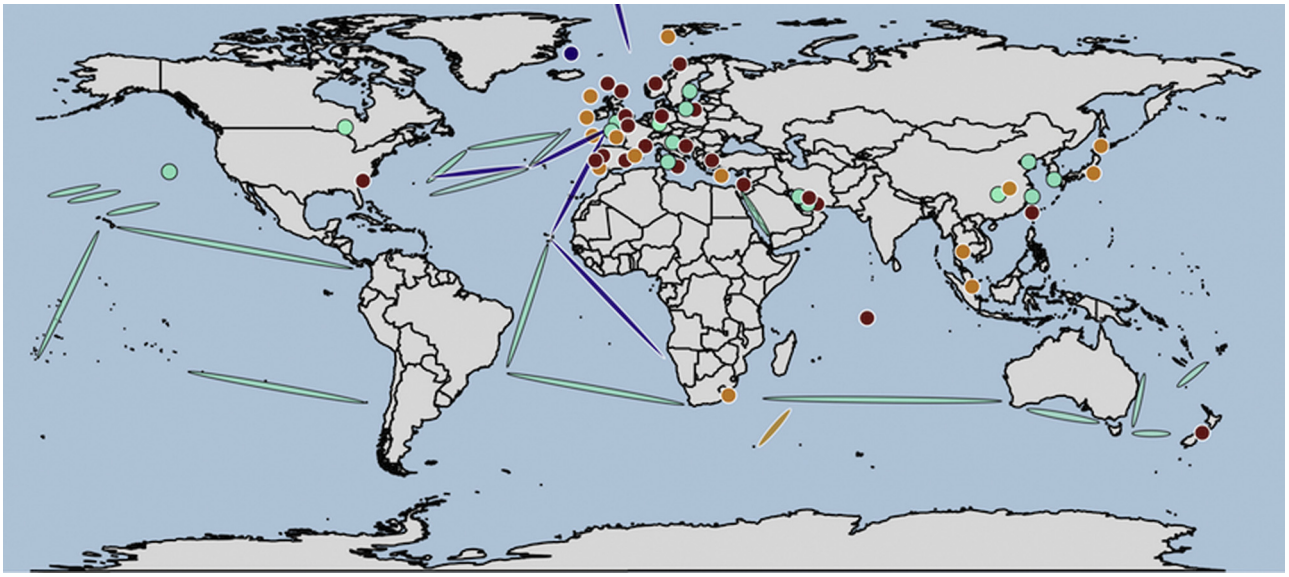

Fig. 2. Sampling sites of studies included in the review [63–91]. Sampling zones are indicated: teal, surface water; dark blue, water column; orange, subtidal and deep sea; red, intertidal. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

sediments (~ 0.1–1 particle m^{-3}), with three exceptions from China and Korea, which presented similar concentrations to intertidal sediments. The data further indicated that subsurface waters contained plastics in similar concentrations to sea surfaces, although the water column was mainly polluted by PP&A (Fig. 4). As previously reported, the concentration of particles detected in deep sea sediments was higher than what was found in intertidal sediments ($> 10^4$ particles m^{-3}; [5]) although, interestingly, no PE or PP were reported in that study.

4. Discussion

Our meta-analysis confirmed that PE, PP, PS and PP&A were among the most abundant polymer types in aquatic environments. This is not surprising as these materials accounted for 74% of global plastic production in 2015 and are commonly used in short life-cycle products [46,47]. Here we show that the relative abundance of low-density polymer types (*i.e.* PE and PP) is highest in open sea surfaces, but lower in intertidal- or subtidal samples, and further decreases in subsurface water (Fig. 5). In turn, plastics denser than seawater, such as polyester, polyamide and acrylics (PP&A), were relatively more abundant in subsurface water than on sea surfaces. The available data from the deep sea further revealed PP&A as the dominant group of plastics in these environments (77%, Fig. 5), as well as chlorinated PE [40], which is similarly dense and mostly added to PVC (ρ ~ 1.16 g cm^{-3}, [48]). Hence, while the relative abundance of different polymers remained similar between shallow intertidal and subtidal sediments, data from the open ocean suggests a segregation between polymers through the water column and does not support the idea that sedimentation is occurring at similar rates for all microplastic polymer types.

If deep sea sediments were the major sink for microplastics in marine systems, we would expect to find throughout the water column similar proportions of polymer types as observed in coastal zones. Moreover, if sinking was a principal mechanism for removal of all types of sea surface plastics, then relative abundance of polymer types should remain similar between surface water and the water column, as well as the deep sea. Particle concentrations supported the trends observed from our prevalence-based analyses (Fig. 4). Given the segregation of different polymer types found here, it is important to reconsider the sink of those plastics "missing" from surface seawater (mainly PE and PP; [3]). Recent studies have already highlighted that up to 97% of this material might have been overlooked, if sampling methods and analysis did not account for vertical mixing [49,50] or if methods for quantifying the smaller fraction of microplastics (*i.e.* < 1 mm) were not implemented [51,52]. Simulations predicted that buoyant PE microparticles would persist and oscillate in the water column [53], but the overall relatively low abundance of PE and PP in studies that have sampled the water column (Figs. 4 and S1-S2), and the lack of evidence for significant plastic debris in sediment traps from the North Atlantic gyre [54], suggest that this issue may be more complex than anticipated. For instance, small buoyant microplastics sink as part of fecal pellets when ingested [28] or when incorporated in marine snow [29,55], but it is uncertain for how long microplastics remain in such particulate organic matter, since (I) fecal pellets containing microplastics are more likely to fragment [28], (II) zooplankton is known to break up larger aggregates [56], and (III) organic material is remineralized in marine aphotic zones [57], altogether perhaps leading buoyant microplastics to reemerge. Deposition of buoyant microplastics may therefore occur more likely in photic-, rather than aphotic sediments, which would stand in agreement with our results (Fig. 5). All of these points, coupled to the analysis we present here, do not allow to conclude that buoyant polymer types reliably sink out of the water column, and emphasize that additional sampling of the deep sea water column and sediments, as well as data from sedimentation traps are necessary to provide an answer to this question.

Unexplained heterogeneity between studies remained very high, despite accounting for sampling depth, highlighting that important moderators remained unaccounted for in the analysis. For instance, local prevalence of specific polymer types may vary with the presence of production plants or specific activities that release characteristic types of polymers (*e.g.* the large number of acrylics found in [45]; Figs. 4 and 5), but it is currently not possible to have higher spatial resolution. Moreover, it is evident that differences between sampling methods, such as the densities employed to extract plastics from sediments or different surface water sampling techniques, can account for variability in results (see Supplementary Information for a detailed discussion). In addition, occurrence and transportation of microplastics in the oceans is subject to wave-driven turbulent mixing [49], as well as plastic properties, such as particle size and shape [22,58]. For instance, unlike other polymers, relative abundance of PS did not significantly change with the sampled zone. This could be explained by the two distinct forms of PS, *i.e.* "solid" PS (ρ ~ 1.04 g cm^{-3}) or expanded PS (ρ < 0.05 g cm^{-3}), which behave differently in water. The very low-

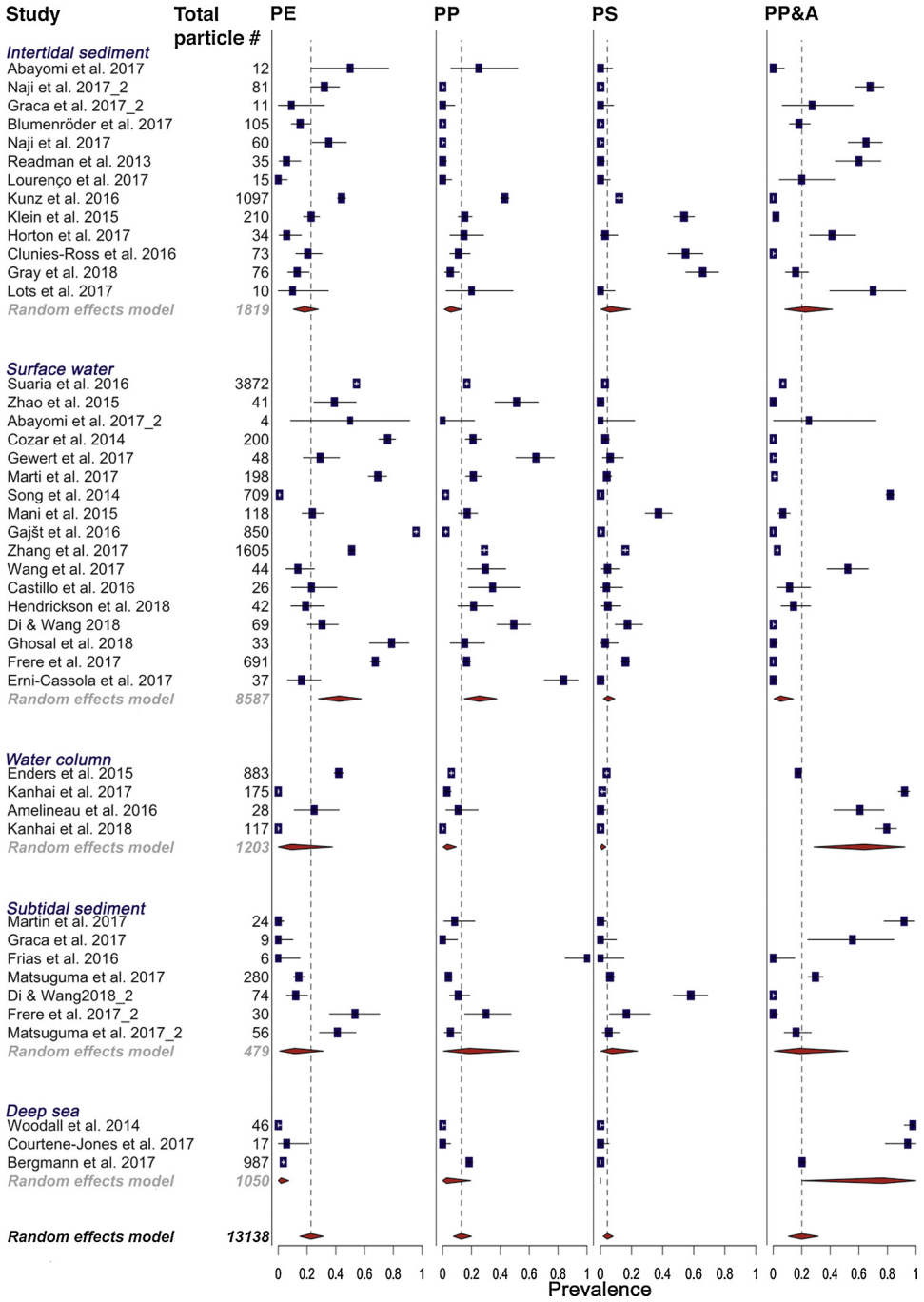

Fig. 3. Prevalence forest plots for analyzed polymer types. For statistical details, including heterogeneity (I^2), see individual polymer type forest plots in supplementary information (figs. S2-S5). Red diamonds represent subgroup means, while the bottommost indicates the overall mean, also represented *via* the dotted line. PE: polyethylene; PP: polypropylene; PS: polystyrene; PP&A: polyester, polyamide and acrylic. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

density form is expected to remain on sea surfaces and intertidal areas (Fig. 4; [59]), whereas the denser form of PS should theoretically sink, thus explaining the occurrence of PS in subtidal samples (7%, Fig. 3).

Undoubtedly, concentration and prevalence of plastic polymer types in aquatic environments vary with geographic location, *i.e.* in response to local sources of pollution, such as densely populated coastal regions [60] and with distance to coast [61], but also within sampling sites [62], the shape of the particles [50], and with the sampling methodology used. While all polymer types can be found in any given sampling zone, our meta-analysis reveals a general trend in relative abundance of four common polymer types in different sampling zones and highlights important knowledge gaps as well as reporting issues. Here, we cannot confirm that buoyant polymer types reliably sink out of the water column, and hence, further research is required to determine the ultimate fate of buoyant plastic polymers such as PE and PP, a fundamental requirement to assess the real risk plastic pollution poses to aquatic life.

Funding

G.E.-C. was supported by a NERC CENTA Ph.D. studentship. J.A.C.-O. was supported by a NERC Independent Research Fellowship NE/

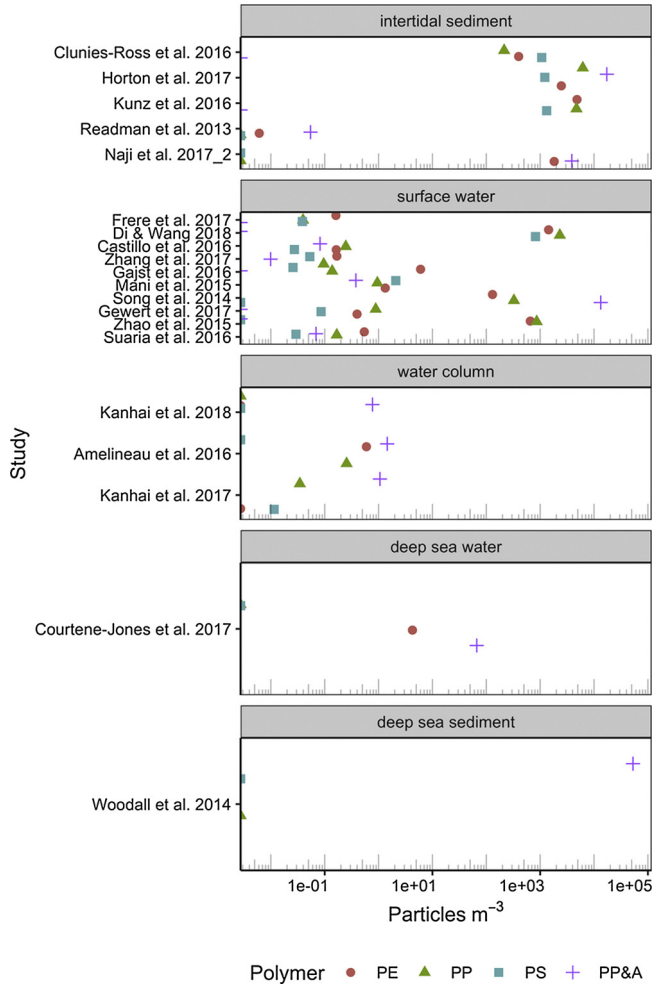

Fig. 4. Concentration of different polymer types within the different sampling zones. PE: polyethylene; PP: polypropylene; PS: polystyrene; PP&A: polyester, polyamide and acrylic.

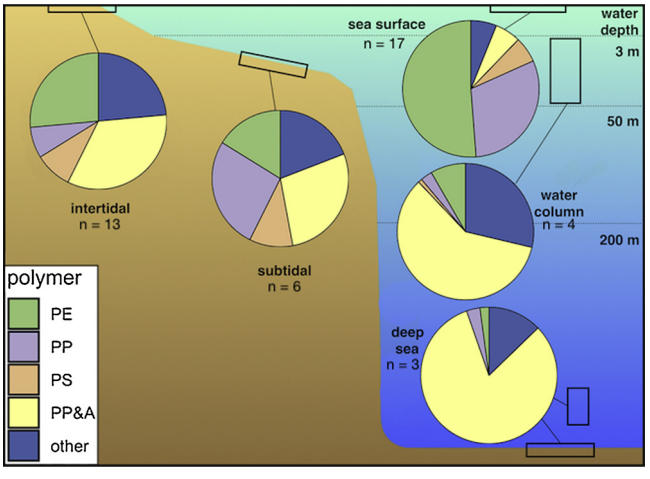

Fig. 5. Relative abundance of common polymer types in different sampling zone. Pie charts represent abundance data normalized to model predictions. For details see forest plots in supplementary information (Figs. S2–S5). PE: polyethylene; PP: polypropylene; PP&A: polyester, polyamide and acrylic; PS: polystyrene. Number of studies in each zone is indicated (n).

K009044/1. V.Z. was supported by CONICYT-BECAS CHILE/Doctorado Becas Chile en el Extranjero, Folio72160583.

Author contributions

G.E.-C. and J.A.C.-O. conceived the study; G.E.-C. reviewed the literature, and G.E.-C. and V.Z. collected the data; G.E.-C. performed the data analysis. G.E.-C. wrote the primary manuscript; J.A.C.-O., V.Z. and M.I.G. provided their expertise and reviewed the manuscript.

Competing interests

The authors declare no competing interests.

Data availability

Original data set and analysis script available from authors upon request.

Novelty statement

Plastic debris is thought to be a hazardous material to marine life, but risk requires evaluating the availability of microplastics to biota and, currently, the ultimate sinks for these materials remains unclear. For the first time, this meta-analysis brings together data recorded from different marine zones and identifies the sinks for microplastics in aquatic systems. Deep sea sediments have been suggested as the final sinks for marine microplastics, but our meta-analysis demonstrates that microplastics segregate through the water column in accordance to the material's density and, hence, final sinks in aquatic environments are different for different polymer materials.

Acknowledgments

The authors thank Robyn J. Wright and Audam Chhun for comments on the manuscript.

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jhazmat.2019.02.067.

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